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We have studied the physics underlying some of the pending issues in quantum cascade lasers, with an emphasis on electronic transport across InAs/AlSb/GaSb heterojunctions – the building block of type II quantum cascade lasers – and on its dependence on the materials' epitaxial growth conditions. We have used resonant tunneling experiments to confirm the coexistence of electrons and holes in separate InAs and GaSb layers, to determine their density and to correlate them to interface smoothness and optimum growth conditions. We have also examined the effectiveness of a superlattice placed on the "collector" end of a type I tunneling heterostructure device to block tunneling through the potential barrier. Finally, we have studied the interaction of photons and excitons in type I microcavities, and have devised a simple way of enhancing the coupling between photons and excitons.

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1. Introduction

The *intersubband* quantum cascade laser[1], which relies on optical transitions between two subbands of a quantum well (or a superlattice) formed by the conduction-band discontinuity between two semiconductors, has opened the door to dependable coherent light sources in a very broad range of wavelengths, from the infrared[2,3] to the terahertz[4] regions of the electromagnetic spectrum. In this new type of lasers an electron generates several photons as it sequentially cascades down pairs of subbands in several heterojunction active regions in "series". Impressive advances in the last few years have led to lasers operating continuously at room temperature[5] and to emission powers of up to several watts[6].

The main drawback of intersubband quantum cascade lasers, based on the InGaAs/InAlAs or the GaAs/GaAlAs system (type I system), is their inefficient conversion of electrical energy into coherent radiation, which is responsible for high threshold currents. Some schemes have been devised to reduce this problem. In one of them, the minigap of a superlattice adjacent to the quantum well prevents tunneling out of the excited state[2]. In another, the unique type II band alignment is used to eliminate tunneling in an interband laser based on InAs/InGaSb, which retains cascading effects while exploiting the interband character to suppress phonon relaxation[7,8]. In yet another scheme, intersubband stimulated emission is achieved in type II InAs/GaSb heterostructures[9-11].

In spite of the hypothetical advantages of type II materials, the state-of-the- art in quantum cascade lasers has been defined by interssubband emission in type I junctions. The relative ease to prepare lattice-matched heterostructures based on them has spurred a large body of work to make them ever more perfect. On the other hand, the greater difficulty to prepare epitaxially Sb-based semiconductors and to lattice match them to substrates or to other materials compatible for type II structures, has made advances in this system much more slower. In addition, some basic parameters (e.g., precise band discontinuities) and the correlation between electronic transport and interface quality are not sufficiently well known.

In this work we have considered the physics underlying some of the pending issues in quantum cascade lasers, with an emphasis on the electronic transport across InAs/AlSb/GaSb heterojunctions – the building block of type II quantum cascade lasers – and on its dependence on the materials' epitaxial growth conditions. We have used extensively resonant tunneling experiments because this physical mechanism is particularly sensitive to interface quality and, in type II structures, to the coexistence of electrons and holes separately in InAs and GaSb. We have also examined the effectiveness of a superlattice placed on the "collector" end of a type I tunneling

heterostructure device to block tunneling through the potential barrier. Finally, in preparation for the day when "vertical cavity quantum cascade lasers" become a reality, we have studied the interaction of photons and excitons in type I microcavities, and have devised a simple way of enhancing the coupling between photons and excitons.

2. Main Results

For clarity, we divide our results along the two categories of materials studied.

2-1. Type II Heterostructures

In InAs/AlSb/GaSb heterojuntions, electrons and holes accumulate at the InAs and GaSb sides of the two material interfaces, respectively. This accumulation is a consequence of the transfer of electrons from GaSb to the conduction band of InAs, whose bottom lies lower in energy than the top of the valence band of GaSb. The thickness of the AlSb layer between InAs and GaSb determines how many electrons are actually transferred. In the absence of extrinsic carriers, the number of electrons in InAs and holes in GaSb should be the same.

We have considered GaSb/AlSb/InAs/AlSb/GaSb and InAs/AlSb/GaSb/AlSb/InAs – two configurations that are particularly suited for elucidating the roles of electrons and holes in transport and the influence of epitaxial-film growth conditions on the physical process itself.

2-1-a. Effect of Growth Temperature

We have studied the tunnel ("vertical") current-voltage (I-V) characteristics of two diodes (A and B) with similar structures [p-GaSb substrate/ AlSb barrier (30 Å to 40 Å)/ InAs quantum well (150 Å to 200 Å)/ AlSb barrier (30 Å to 40 Å)/ p-GaSb counterelectrode] but grown at different temperatures. Diode A was prepared at 500 C and diode B at 400 C, both at the Army Research Laboratory. The latter was prepared under similar growth conditions as ARL's most efficient interband quantum cascade lasers.

At room temperature, the I-V. characteristic of diode A exhibited a well-developed negative differential resistance (NDR), but that of diode B did not. At low temperature (T = 77K) both diodes showed good NDR but the "valley" current of diode B was much larger than that of diode A. Although "leakage" currents can arise from diode-processing processing, this possibility was practically ruled out here as the results were identical for different processing methods. It is much more likely that in this case the leakage is a consequence of "internal" leakage paths created by defects formed during the growth of the heterostructure.

Conclusive evidence of the lower quality of diode B was provided by the zero-voltage conductance of the I-V characteristics when the sample was at low temperature (T = 4K) in a magnetic field parallel to the tunnel current direction. In both diodes the conductance oscillated with varying strength of the field H, the oscillations being periodic with the inverse of the field and from which it is straightforward to get the two-dimensional (2D) electron density in the InAs layer. The density thus deduced was 1.3×10^{16} m^{-2} for diode A and 3.9×10^{16} m^{-2} for diode B. While the former value is typical for 150 Å – 200 Å wide InAs wells, the latter is unusually large and found only in

low-quality heterostructures. The conductance oscillations appeared in diode B at much higher magnetic fields than in diode A. Since the onset field of the oscillations is directly related to the electronic mobility we inferred that the mobility of diode B was about an order of magnitude lower than that of diode A, or about 3 m^2 /Vs.

Based on these results, we conclude that although the quality of the low-temperature heterostructures could have been quite good from the point of view of optical emission it was relatively poor for vertical transport, which raises the question of whether there is a correlation between the two physical processes, and, if so, whether it is possible to improve the epitaxial growth so that more efficient optical transitions are achieved.

2.1-b. Magnetotunneling: In-plane Field

We have selected the highest quality GaSb/AlSb/InAs/AlSb/GaSb heterostructures for vertical transport experiments in the presence of a magnetic field parallel to the interfaces. The goal was to establish unequivocally the presence of spatially-separated two-dimensional (2D) electrons and holes. Such a bi-layer system is of interest because, among other things, it offers the possibility of forming a gas of excitons aligned perpendicularly to the interface planes, a configuration that, according to theoretical predictions, may lead to Bose-Einstein condensation-like phenomena.

We have found that at a temperature of 4.2K the zero-bias tunneling conductance exhibits a very rapid increase with increasing field, peaks at 5.8T, and then gradually declines except for a secondary peak at 16.3T. The presence of two clear peaks in the magnetoconductance is an unambiguous indication of the existence of 2D holes in the GaSb electrodes. At the same time, the values of the magnetic field at which the two peaks were observed have allowed us to determine the ratio of 2D electron to hole densities, as well as the effective separation between these two gases[12].

We have found the 2D electron and hole densities to be $N_e = 1.2\times10^{12}$ cm^{-2} (in the InAs central well) and $N_h = 2.7\times10^{11}$ cm^{-2} (in <u>each</u> GaSb interface), respectively. That the N_e/N_h ratio is much larger than the ideal value of 2 (since the total number of electrons and holes is the same) confirms the existence of extrinsic sources of electrons in InAs/AlSb heterostructures[13,14]. For the effective separation between the 2D electrons in InAs and the 2D holes at the GaSb interfaces, we have found a value of 164 Å. This number is to be compared with the geometrical distance of 115 Å between the center of the InAs well and the AlSb/GaSb interface. The difference between these two figures represents the average distance of the one-dimensional hole wavefunction to that physical interface.

To summarize this section, from the study of the magnetotunneling current in GaSb-AlSb-InAs-AlSb-GaSb heterostructures we have shown the 2D character of the holes that participate in the tunneling process and determined the small effective separation between 2D electrons and holes in these structures. In contrast with these encouraging results, the charge imbalance we have found hinders electron-hole binding and the formation of a gas of oriented excitons with opposite charges in different planes.

2.1-c. Magnetotunneling: Perpendicular Field

The unusual energy-band alignment of heterostructures that combine InAs and GaSb has made them attractive not only for quantum-cascade lasers but also for spin devices, in which the electron's spin states, rather than its charge, determine the operation of the device, be it a transistor or a resonant-tunneling device[15,16]. On the road to practical heterostructure-based spin devices, one of the first steps has been to establish the presence of Rashba splitting and to determine its amount, and, if possible, to control it. This effect refers to the splitting and uneven population of the two spin subbands in the conduction band of a semiconductor for non-zero in-plane wave vectors, in the absence of an external magnetic field.

Since some of the proposed spin devices rely on electrons tunneling across a heterostructure, it is important to sort out effects in vertical transport that might be construed as evidence of spin splitting. To this effect, we have done a systematic magnetotunneling study using a variety of InAs-AlSb-GaSb-AlSb-InAs structures with different thickness for the GaSb central well and the AlSb barriers[17,18]. In all the structures, 2D electron gases accumulated in the quasi-triangular wells formed in the conduction band of InAs at the InAs-AlSb interfaces. From a comparison and analysis between our experimental results and a calculation of the magnetoconductance we conclude that the two separate sets of oscillations in the conductance that we have observed are not a consequence of Rashba splitting, as might be inferred from a superficial analysis, but are rather the result of an asymmetry between the two interfaces[18].

With the magnetic field applied perpendicularly to the materials interfaces, the low-temperature (T = 1.7K) tunneling conductance at zero bias exhibited Shubnikov-de Haas oscillations. In contrast with similar measurements in heterostructures in which InAs constituted the central well rather than the end electrodes, the structures with GaSb in the center showed clear beating effects in the amplitude of the oscillations, which suggested two different electron populations, possibly as a consequence of Rashba splitting – or of some other mechanism.

To discern among several possibilities, we have used a simple model to calculate the magnetoconductance, which we have then compared with the experimental results. The basic assumptions of our model are that the oscillations are only due to electrons in the two InAs triangular wells and that there is a small energy difference between the quantized states in the wells. From a comparison between theory and experiment we have confirmed that indeed there is a splitting of quantum states, and, equivalently, two different electron populations. But, most important, the splitting and carrier differences vary widely across the heterostructures, even though their material parameters were not much different from one to another. We have therefore concluded that it is the carrier densities in the two InAs wells that are different, not that there are two different populations in each well, as Rashba splitting would imply. The difference in carrier density depends on the epitaxial-growth conditions, which determine the effective difference between the two InAs interfaces.

2.1-d. Effect of Interface Smoothness

The magnetotunneling experiments described in 2.1-c have allowed us to extract valuable information about the optimal growth conditions to obtain smooth interfaces in InAs-AlSb-GaSb heterostructures. Two structures with the same composition and layer thickness, and grown under optimal conditions to form an InSb-type bond at the AlSb-InAs interfaces, showed nevertheless quite different magnetotransport behavior. Although the quality of both structures was excellent (based on the small magnetic field at which the conductance oscillations first become visible), only one of them showed no trace of beating effects, in other words, identical InAs interfaces. The only controlled difference between the two epitaxial growths was that the sample with electrically identical interfaces was prepared by adding an extra ¼ In monolayer on the bottom AlSb-InAs interface. This extra layer apparently reduces interface roughness even further, as revealed by plan-view scanning tunneling microscopy[19,20].

These results are very encouraging in that they show that, under optimum growth conditions, balanced populations of high-mobility 2D electron gases can be formed in InAs accumulation layers, with which 2D-2D interaction effects can be probed.

2-2. Type I Heterostructures

Intersubband quantum cascade lasers have been realized in two families of type I heterostructures: GaAs-GaAlAs and InGaAs-InAlAs. The former one has the advantage that the quantized states are formed in a binary compound (free of alloy scattering); on the other hand, in the latter the potential barrier that confines those states is higher, thus reducing detrimental out-of-state tunneling. We have focused on heterostructures based on GaAs-GaAlAs because of the easier epitaxial growth conditions for lattice-matched multilayers.

2.2-a. Tunnel-blocking Effects of a Superlattice

Essential to the effective operation of quantum-cascade lasers is the ability to reduce (if not eliminate) out-of-the-well tunneling of electrons in an upper level of the well, before they relax to a lower level and emit radiation. In type I cascade lasers this is achieved by a superlattice placed next to the "exit" barrier of each active section of the laser. The "mini-bandgap" between superlattice subbands blocks electrons attempting to tunnel out of the well and makes the radiation process more efficient. In this scheme tunneling is prevented because of parallel (to the interfaces) momentum conservation, so the question arises, how effective is this blocking mechanism, especially at room temperature. This question is quite pertinent, in view of the well-known difficulty to achieve negative-differential conductance at room-temperature in GaAs-GaAlAs resonant-tunneling diodes, which are based on the same momentum-conservation principle.

To address that question we have studied the temperature dependence of the current-voltage characteristics of "superlattice-barrier-superlattice" GaAs-$Ga_{0.7}Al_{0.3}As$ heterostructures. Such a configuration simulates the tunneling-blocking process that occurs in cascade lasers. When a voltage difference is applied between the two (doped) superlattice electrodes, electrons from the

emitter tunnel through the barrier to the collector, provided the emitter's and collector's superlattice minibands (width D) have some energy overlap. But if the voltage difference is larger than a critical voltage (= D/ e), beyond which the emitter's electrons face the collector minigap, then tunneling cannot occur -if parallel momentum is to be conserved. As a result, the I-V characteristic shows a region of negative-differential conductance beyond that critical voltage.

In our case, each superlattice electrode consisted of 50 periods of alternating GaAs (42Å) and $Ga_{0.7}Al_{0.3}As$ (20Å) layers, doped uniformly to either 5×10^{16} cm^{-3} or 1×10^{17} cm^{-3} or 1×10^{18} cm^{-3} carriers. The barrier between the electrodes was typically 150Å. In all three cases (of doping) the heterostructures showed I-V characteristics with distinct negative-differential-conductance at T = 77K, typically, with a peak-to-valley around 5. (No much difference was found at T = 4K). However, when the temperature was increased above liquid nitrogen the NDC gradually diminished until it disappeared and the I-V became quasi-ohmic, in some cases, at temperatures as low as 200K (and in all cases at room temperature).

This result shows that, as in resonant-tunneling diodes, temperature can break the conservation of parallel momentum and indirectly facilitate tunneling, thus casting a shadow on the use of a superlattice to block undesirable electron tunneling in cascade lasers at high temperatures.

2.2-b. Tuning of Rabi Splitting in a Superlattice-Microcavity System

One of the most heavily studied aspects of semiconductor microcavities has been the coupling between the excitonic modes of two-level atom-like structures embedded within the cavity region, such as quantum wells, and the photonic mode of the cavity. When large enough, the coupling manifests itself as a Rabi splitting between the two resonant, interacting modes, forming a so-called cavity-polariton[21,22]. One of the main interests has been to enhance that splitting, by increasing either the oscillator strength or the quantum well density. We have demonstrated that it is possible to enhance both of these characteristics by embedding a superlattice into a microcavity, subjected to an external electric field[23]; moreover, we have shown that the magnitude of the Rabi splitting can be tuned with an electric field[24]. These new results, which hinge on our demonstration, several years ago, of field-induced Wannier-Stark localization of superlattice states[25], could have applications in the development of opto-electronic devices.

By embedding a GaAs-GaAlAs superlattice in a semiconductor microcavity subjected to an electric field we have controlled the exciton-photon interaction and drastically modified the Rabi splitting. The superlattice fills the entire cavity, thus increasing the exciton density, while the oscillator strength of the superlattice exciton is enhanced by the Stark localization induced by the electric field. In this way, we have been able to almost double the Rabi splitting, from 6.5 meV, typically observed in conventional GaAs-GaAlAs microcavities, to 11.5 meV (Ref. 23). Moreover, by varying the degreee of exciton localization through a controlled change of the field, we have tuned the Rabi splitting from 5 meV, when the field is zero, to 11.5 meV for an optimum field of 30 kV/cm (Ref. 24). Beyond this field the Rabi splitting gradually decreases as a result of the quantum-

confined Stark effect in isolated quantum wells, which decreases the exciton's oscillator strength and shifts its energy away from resonance with the cavity.

As a result of our work, the enhanced Rabi splitting can now be easily observed at room temperature; besides, it will be possible to use our scheme for modifying the exciton-photon coupling in studies (e.g., time-resolved spectroscopy) aimed at elucidating this interaction or in devices exploiting it.

2.2-c. Shot Noise in Negative-Differential-Conductance Devices

To gain a deep insight on the transport mechanisms of an electronic or an optoelectronic device it is sometimes essential to measure its shot noise, as dramatically illustrated by this work. We have compared two types of semiconductor devices that show the same current-voltage (I-V) characteristics and yet their current transport mechanisms are very different. We have observed that the shot noise of a double-barrier resonant tunneling diode is partially suppressed relative to Poisson noise ($S = 2eI$) in the linear region of its I-V characteristics and enhanced in the negative-differential-conductance region[26,27]. In sharp contrast, we have found that the shot noise of a single-barrier tunneling diode with electrodes made out of doped superlattices is Poissonian throughout the entire I-V characteristics[27]. Our result implies that charge accumulation, not just the system instability that follows negative differential conductance, is responsible for shot-noise enhancement.

Our work has proved wrong a recent theoretical prediction that system instability, not charge accumulation, is essential to enhancement of shot noise in negative-differential-conductance devices. The work also highlights a simple way of discerning between competing transport mechanisms in a device whose I-V characteristics are known but whose microscopic transport processes are still in question. This is particularly valid for the case of quantum-cascade lasers, whose building blocks are closely related to the simple devices studied here.

3. Publications and Conference Presentations

Peer-reviewed journals:

1. *Optical Gain of Type II based Quantum Cascade Lasers*, J. L. Jimenez and E. E. Mendez, Solid State Commun. **110**, 537 (1999).
2. *Magnetotunneling in a Two-Dimensional Electron-Hole System Near Equilibrium*, E. M. Gonzalez, Y. Lin, and E. E. Mendez, Phys. Rev. B **63**, 033308 (2000).
3. *Ultrafast Light-Polarization Dynamics in Semiconductor Microcavities*, M. D. Martín, L. Viña, and E. E. Mendez, Solid State Comm. **119**, 259 (2001).
4. *Enhancement of Rabi Splitting in a Microcavity with an Embedded Superlattice*, J. H. Dickerson, E. E. Mendez, A. A. Allerman, S. Manotas, F. Agulló-Rueda, and C. Pecharromán, Phys. Rev. B **64**, 155302 (2001).
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6. *Electric Field Tuning of the Rabi Splitting in a Superlattice-Embedded Microcavity*, J. H. Dickerson, J. K. Son, E. E. Mendez, and A. A. Allerman, Appl. Phys. Lett. **81**, 803 (2002).
7. *Tunneling Characteristics of an Electron-Hole Trilayer in a Parallel Magnetic Field*, Y. Lin, E.E. Mendez, and A.G. Abanov, Phys. Rev. B. **66**, 195311 (2002).
8. *Shot Noise in Negative-Differential-Conductance Devices*, W. Song, E. E. Mendez, V. V. Kuznetsov, and B. Nielsen, Appl. Phys. Lett. **82**, 1568 (2003).
9. *Magnetotunneling between Two-dimensional Electron Gases in InAs-AlSb-GaSb Heterostructures*, Y. Lin, E. M. Gonzalez, E. E. Mendez, R. Magno, B. R. Bennett, and A. S. Bracker, Phys. Rev. B **68**, 035311 (2003).

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1. *Anomalous Resonant-tunneling Effect in Type II Heterostructures*, E. E. Mendez, V. V. Kuznetsov, D. Chokin, and J. D. Bruno, Physica E **6**, 335 (2000). 13th Int. Conf. Electron. Properties of Two-dimensional Systems, Ottawa, Canada, Aug.2-6, 1999.
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Conference presentations:

1. *Magnetotunneling Between Two InAs Quantum Wells*, A. Sacedon, E. M. Gonzalez, Y. Lin, and E. E. Mendez, American Physical Society Annual Meeting, Minneapolis, March 20-24, 2000.
2. *Tunneling Between Two Dimensional Electron and Hole Gases*, Y. P. Lin, E. M. Gonzalez, and E. E. Mendez, American Physical Society Annual Meeting, Seattle, March 12-16, 2001.
3. *Enhanced Rabi Splitting in a Superlattice-embedded Microcavity*, J. Dickerson, E. E. Mendez, S. Manotas, F. Agullo-Rueda, and A. A. Allerman, American Physical Society Annual Meeting, Seattle, March 12-16, 2001.
4. *Enhanced Rabi Splitting in a Microcavity-Superlattice Sytem*, J. Dickerson, E. E. Mendez, S. Manotas, F. Agullo-Rueda, and A. A. Allerman, *Type II Materials and Characterization*, E. E. Mendez, Quantum Cascade Laser Workshop, Arlington, VA, October 9, 2001.
5. *Shot Noise in Negative Differential Conductance Devices*, (Invited) E. E. Mendez, NATO Workshop on Quantum Noise in Mesoscopic Systems, Delft, The Netherlands, June 2-4, 2002.

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